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(54) Title: ELASTIC MATERIALS AND ARTICLES THEREFROM

(57) Abstract

Elastic materials, particularly films, strips, coatings, ribbons and sheets made from at least one substantially linear ethylene polymer are disclosed which can be fabricated on conventional polyolefin extrusion equipment. They can be used to produce elastic composite structures that are stretchable and have recycle compatibility between elastic and non-elastic components. The novel materials have at least about 80 percent recovery at 100 percent strain and are especially useful in making fabricated composite articles (e.g., disposable diapers).

ELASTIC MATERIALS AND ARTICLES THEREFROM

This invention relates to elastic materials, particularly, elastic

5   films, strips, sheets and composites, and to articles fabricated
therefrom, such as disposable incontinence garments or diapers. The
elastic materials comprise at least one judiciously selected homogeneously
branched substantially linear ethylene polymer.

10   Materials with excellent stretchability and elasticity are needed to
manufacture a variety of disposable and durable articles, such as
incontinence garments, disposable diapers and furniture upholstery.
Stretchability and elasticity are desirable characteristics to effectuate
a closely conforming fit to the body of the wearer or to the frame of the

15   item. It is also desirable to maintain the conforming fit during repeated
use, extensions and retractions. For incontinence articles,
stretchability and elasticity are particularly desirable to insure comfort
and provide security against unwanted leaks.

20   Disposable articles are typically prepared by the combination of
polymer fibers, films, sheets and absorbent materials. Whereas the fibers
are prepared by well known processes such as spunbonding, melt blown and
continuous filament wounding, the film and sheet forming processes
typically involve known extrusion and coextrusion processes, for example,

25   blown bubble extrusion, extrusion casting, profile extrusion, injection
molding, extrusion coating and extrusion sheet. The resultant elastic
film, coating or sheet may be subsequently cut or slit to short lengths
and/or narrow widths to prepare strips, tapes, bands, ribbons or the like.

30   There are at least two ways elastic materials are employed to
manufacture disposable and durable articles. Elastic films, strips and
sheets are used as uncombined elastic components (panels or portions), or
they are constructed as or into multilayer structures to provide elastic
composite materials with enhanced elasticity and stretchability. In a

35   diaper, for example, experimental and commercial uses include in or as
side panels, waist bands, backsheets, leg bands, and even topsheets where
the elastic material is rendered pervious or "breathable" by such methods

as apperturing, slitting, or microperforating as suggested by Lippert et al. in USP 4,861,652.

An example of the use of elastic materials to construct elastic composite materials is provided by Van Gompel et al. in USP 4,940,464, USP 4,938,757 and USP 4,938,753. Van Gompel et at. disclose disposable incontinence garments containing elastic gathering means and stretchable side panels. The gathering means and stretchable side panels are made from film of block or graft copolymers such as butadiene, isoprene, styrene, ethylene-methyl acrylate, ethylene-vinyl acetate, ethylene-ethyl acrylate or blends thereof.

An example of use of elastic materials to construct composites with the particular benefit of enhanced stretchability is a stretchable fastening tape for a disposable diaper disclosed by Gesp in USP 5,057,097.

There has been a persistent need for extrudable materials suitable for producing films, strips, sheets and composites with excellent stretchability and elasticity. Although there are a variety of elastic materials currently available, these known solutions require blending or additive incorporation to meet desire levels of extrusion processability, stretchability or elasticity. Still other proposed solutions such as the method disclosed by Butin in USP 3,849,241 require 'controlled thermal and oxidative degradation' of the elastic material to affect viscosity adjustments prior to extrusion. Yet other prior art solutions, such as the disclosure by de Marais in USP 4,323,534 requires a post-extrusion leaching step to extract processing aids such as fatty acid amides. Moreover, prior art elastic materials generally involve expensive elastomers such as styrene butadiene copolymers, polyether block amides, polyether esters and polyurethanes which typically necessitate blending with polyolefins for adequate extrusion processability.

Where polyolefins themselves have been previously employed as elastic materials, other problems have arisen. For example, where ethylene/α,β-unsaturated copolymers are known to possess improved elasticity as a function of increased comonomer levels, Daponte in USP

4,803,117 discloses ethylene vinyl ester copolymers with high vinyl ester levels are requisite to effectuate adequate elasticity for disposable articles. However, these high vinyl ester levels invariably renders the polymer susceptible to undue thermal degradation.

Several definitions have been adopted herein as previously described by Collier et al. in USP 5,169,706. The term "elastic" means that the film will recover at least about 50 percent of its stretched length after the first pull and after the fourth pull to 100 percent strain (doubled the length). Elasticity can also be described by the "permanent set" of the film. Permanent set is the converse of elasticity. A film is stretched to a certain point and subsequently released to the original position before stretch, and then stretched again. The point at which the elastic material begins to pull a load is designated as the percent permanent set.

The term "stretchable" is used herein in reference any material which, upon application of a biasing force, elongates at least about 60 percent (that is, to a stretched, biased length which is at least about 160 percent of its relaxed unbiased length), and which, will recover at least 55 percent of its elongation upon release of the stretching, elongating force. A hypothetical example would be a one (1) inch sample of a material which is elongated or stretched to at least 1.60 inches (4.06 cm) and which, upon being elongated to 1.60 inches (4.06 cm) and released, will recover to a length of not more than 1.27 inches (3.23 cm). Many elastic materials may be elongated by much more than 60 percent (that is, much more than 160 percent of their relaxed length), for example, elongated 100 percent or more, and many of these will recover to substantially their initial relaxed length, for example, to within 105 percent of their original relaxed length, upon release of the stretching force.

As used herein, the terms "recover" and "recovery" refer to a contraction of a stretched material upon termination of a biasing force following stretching of the material by application of the biasing force. For example, if a material having a relaxed, unbiased length of one (1)

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inch is elongated 50 percent by stretching to a length of one and one half (1.5) inches (3.81 cm) the material would be elongated 50 percent (0.5 inch) (1.27 cm) and would have a stretched length that is 150 percent of its relaxed length.   If this exemplary stretched material contracted, that is recovered to a length of one and one tenth (1.1) inches after release of the biasing and stretching force, the material would have recovered 80 percent (0.4 inch) (1.02 cm) of its one-half (0.5) (1.27 cm) inch elongation.   Recovery may be expressed as [(maximum stretch length - final sample length) / (maximum stretch length - initial sample length)] x 100.

As used herein, the term "nonelastic" refers to any material which does not fall within the definition of "elastic" or "stretchable" above.

As used herein, the term "less-elastic" includes "nonelastic" and any material referenced apposite an "elastic material".

The term "elastic material" as used herein refers to the films, strips, coatings, tapes, webs, ribbons, bands, and sheets as well as the "elastic composite materials" disclosed herein unless specifically distinguished as pertaining to the prior art.

The term "composite elastic material" as used herein refers to a multilayer material (including a fabric) having at least one elastic material layer joined to at least one less-elastic layer at least at two locations in which the less-elastic layer is extended or gathered between the locations where it is joined to the elastic layer.   A composite elastic material may be stretched to the extent that the less-elastic material between the bond locations allows the elastic material to elongate or stretch.   An example of this type of composite elastic material is disclosed, for example, by U.S. Pat. No. 4,720,415 to Vander Wielen et al., issued Jan. 19, 1988, which is hereby incorporated by reference.

The term "article" as used herein refers to fabricated composite items comprising elastic materials disclosed herein.   Articles include

disposable infant care and adult incontinence care items such as
incontinence garments, training pants and diapers.

5   The term "stretch-to-end" as used herein refers to a ratio
determined from the difference between the unextended dimension of a
composite elastic material and the maximum extended dimension of a
composite elastic material (i.e., additional extension would rupture bonds
which connecting individual layers or components of the composite
together) and dividing that difference by the unextended dimension of the
10   composite elastic material. If the stretch-to-end is expressed as a
percentage, this ratio is multiplied by 100. For example, a composite
elastic material having an unextended length of 5 inches (12.7 cm) and a
maximum extended length of 10 inches (25.4 cm), has a stretch-to-end of
100 percent. Stretch-to-end may also be referred to in prior art as
15   "maximum nondestructive elongation".

As used in the claims herein, the term "consisting essentially
of" means that the substantially linear ethylene polymers used to make the
elastic materials can have additional materials which do not materially
20   affect its elasticity.

The terms "homogeneous branching distribution" and
"homogeneously branched" are defined herein to mean that (1) the α-olefin
comonomer is randomly distributed within a given molecule,
25   (2) substantially all of the copolymer molecules have the same ethylene-
to-comonomer ratio, (3) the polymer is characterized by a narrow short
chain branching distribution where the short chain branching index
(defined herein below) is greater than 30 percent, more preferably greater
than 50 percent, and (4) the polymer essentially lacks a measurable high
30   density (crystalline) polymer fraction as measured by known fractionation
techniques such as, for example, a method that involves polymer fractional
elutions as a function of temperature.

The term "interpolymer" is used herein to indicate a
35   copolymer, or a terpolymer, or the like, where, at least one other
comonomer is polymerized with ethylene to make the interpolymer.

The term "substantially linear" means that the polymer backbone is substituted with 0.01 long chain branch/1000 carbons to 3 long chain branches/1000 carbons, more preferably from 0.01 long chain
5   branch/1000 carbons to 1 long chain branch/1000 carbons, and especially from 0.05 long chain branch/1000 carbons to 1 long chain branch/1000 carbons.

The term "long chain branching" is defined herein as a chain
10   length of at least about 6 carbons, above which the length cannot be distinguished using ^{13}C nuclear magnetic resonance spectroscopy, yet the long chain branch can be about the same length as the length of the polymer back-bone.

15   Long chain branching is determined by using ^{13}C nuclear magnetic resonance (NMR) spectroscopy and is quantified using the method described by Randall (Rev. Macromol. Chem. Phys., C29, V. 2&3, p. 285-297).

20   We have discovered new elastic materials which do not need additives to be elastic. These new elastic films, strips, coatings, ribbons and sheets can be produced on conventional extrusion equipment (for example, blown bubble, extrusion casting, extrusion coating and sheet extrusion) and they can be used to produce highly elastic composites whereby there is
25   excellent recycle compatibility between the elastic and less-elastic components of fabricated disposable articles, particularly when the article comprises only polyolefin materials and absorbent hydrogel materials such as polyacrylic acid supplied.

30   Preparation of the elastic materials disclosed herein does not involve "controlled degradation" prior to extrusion, or a post-extrusion additive extraction step, nor do these elastic materials show any particular susceptibility to undo thermal degradation.

35   These elastic materials are made from novel elastic homogeneously branched substantially linear ethylene polymers which exhibit extrusion

processability characteristics that are substantially improved in contrast
to heterogeneously branched linear low density polyethylene. The benefit
of improved processability is manifested on conventional polyolefin
extrusion equipment as higher extrusion outputs (productivity) and/or as
5   lower die pressure and extruder amperage (energy consumption).

The new elastic materials of the present invention have permanent
set values of about 20 percent or less, or, stated another way, percent
recovery values of at least about 80 percent. This property is
10 surprisingly different than the response of films, strips, coatings and
sheets made from other polyolefins which are known to vary predictably as
a function of polymer density or crystallinity wherein higher density or
crystallinity yields higher percent permanent set values characteristic of
the material being less-elastic or nonelastic. Surprisingly, the elastic
15 substantially linear ethylene polymers disclosed herein are distinguished
from other substantially linear ethylene polymers by their tendency to
show enhanced elasticity as film thickness decreases while other
substantially linear ethylene polymers with the polymer densities greater
than about 0.89 g/cm^3 tend to show equivalent or less elasticity at
20 reduced film thicknesses.

The new elastic materials disclosed herein are made from at least
one homogeneously branched substantially linear ethylene polymer having:

25   a) a melt flow ratio, I_{10}/I_2, $\geq$ 5.63,

            b) a molecular weight distribution, M_w/M_n, defined by the
               equation: $M_w/M_n \leq (I_{10}/I_2)$ - 4.63,

30   c) a critical shear rate at onset of surface melt fracture of
               at least 50 percent greater than the critical shear rate
               at the onset of surface melt fracture of a linear ethylene
               polymer having about the same I_2 and M_w/M_n, and

35   d) a density less than about 0.89 g/cm^3.

Figure 1 graphically displays percent permanent set for 1.5 mil film made from elastic substantially linear ethylene polymers of the present invention versus the density of the polymer as well as three assigned regions of relative elasticity. The curve fit is the result of computer-generated third order polynominal regression analysis and the region assignments are the result of discernible and substantial slope changes within those regions.

Figure 2 graphically displays the logarithmic curve fit for the percent permanent set of film made from both elastic substantially linear ethylene polymers of the present invention and higher density substantially linear ethylene polymers versus film thickness.

Figure 3 is a perspective view of an elastic composite material of the present invention with enhanced stretchability comprising an elastic material layer attached to a nonelastic material layer; the elastic material layer is disclosed herein.

Figure 3A is a top plan view of the inventive elastic composite material of Figure 3.

The substantially linear ethylene polymers used in the present invention are a unique class of compounds that are further defined in copending application serial number 07/776,130, now U.S. Patent No. 5,272,236, filed October 15, 1991 and in copending application serial number 07/939,281, now U.S. Patent No. 5,278,272, filed September 2, 1992. The teachings disclosed in these copending applications include the utilization of constrained geometry catalysts and suitable methods of production.

The homogeneously branched substantially linear ethylene polymers are characterized as having:

      a) a melt flow ratio, I_{10}/I_2, ≥ 5.63,

      b) a molecular weight distribution, M_w/M_n, defined by the equation: $M_w/M_n \leq (I_{10}/I_2) - 4.63$, and

c) a critical shear stress at onset of gross melt fracture
greater than 4×10^6 dyne/cm^2.

5   The homogeneously branched substantially linear ethylene polymers
can also be characterized as having:

a) a melt flow ratio, I_{10}/I_2, ≥ 5.63,

b) a molecular weight distribution, M_w/M_n, defined by the
10   equation: $M_w/M_n \leq (I_{10}/I_2) - 4.63$, and

c) a critical shear rate at onset of surface melt fracture at
least 50 percent greater than the critical shear rate at
the onset of surface melt fracture of either a
15   homogeneously or heterogeneously branched linear ethylene
polymer having about the same I_2 and M_w/M_n.

The novel homogeneously branched substantially linear ethylene
polymers used to make the elastic materials are easily distinguished from
20 homogeneously branched linear ethylene polymers. The term "homogeneously
branched linear ethylene polymer" means that the polymer does not have
long chain branching. That is, the linear ethylene polymer has an absence
of long chain branching, as for example the traditional heterogeneously
branched linear low density polyethylene polymers or linear high density
25 polyethylene polymers made using Ziegler polymerization processes (for
example, USP 4,076,698 (Anderson et al.) or linear homogeneously branched
polymers (for example, USP 3,645,992 (Elston). The term "linear ethylene
polymers" does not refer to high pressure branched polyethylene,
ethylene/vinyl acetate copolymers, ethylene/acrylic acid copolymers, or
30 ethylene/vinyl alcohol copolymers which are known to those skilled in the
art to have numerous long chain branches.

The homogeneously branched substantially linear ethylene polymers
used to form the elastic film, strips, coatings, ribbons and sheets of the
35 present invention have homogeneous branching distributions and a single
melting peak, as measured using differential scanning calorimetry (DSC),

in contrast to heterogeneously branched linear ethylene polymers, which
have 2 or more melting peaks, due to the heterogeneously branched
polymer's broad short chain branching distribution.

5   A unique characteristic of the homogeneously branched substantially
linear ethylene polymers used to prepare the elastic films, strips,
coatings, ribbons and sheets of this invention is a highly unexpected flow
property where the I_{10}/I_2 value of the polymer is essentially independent
of polydispersity index (i.e. M_w/M_n) of the polymer. This is contrasted
10   with conventional linear homogeneously branched and linear heterogeneously
branched polyethylene resins having rheological properties such that to
increase the I_{10}/I_2 value, the polydispersity index must also be
increased. In addition, the novel homogeneously branched substantially
linear ethylene polymers of this invention also exhibit enhanced
15   processability as manifested by a higher extrusion output, lower pressure
drop through screen packs and gel filters, lower extruder amperage and
lower die pressure.

        Suitable unsaturated comonomers useful for polymerizing with
20   ethylene to prepare the elastic materials of this invention include, for
example, ethylenically unsaturated monomers, conjugated or non-conjugated
dienes, polyenes, etc. Examples of such comonomers include C_3-C_{20}
α-olefins as propylene, isobutylene, 1-butene, 1-hexene, 4-methyl-1-
pentene, 1-heptene, 1-octene, 1-nonene, and 1-decene. Preferred
25   comonomers include propylene, 1-butene, 1-hexene, 4-methyl-1-pentene and
1-octene, and 1-octene is especially preferred. Other suitable monomers
include styrene, halo- or alkyl-substituted styrenes, tetrafluoroethylene,
vinylbenzocyclobutane, 1,4-hexadiene, 1,7-octadiene, and cycloalkenes, for
example, cyclopentene, cyclohexene and cyclooctene.

30

        The density of the homogeneously branched substantially linear
ethylene polymers used to make the elastic materials of the present
invention is measured in accordance with ASTM D-792 and is generally less
than about 0.89 g/cm^3, preferably from 0.85 g/cm^3 to 0.89 g/cm^3, and
35   especially from 0.85 g/cm^3 to 0.88 g/cm^3.

The molecular weight of the substantially linear ethylene interpolymers is conveniently indicated using a melt index measurement according to ASTM D-1238, Condition 190 C/2.16 kilogram (kg), formerly known as "Condition E" and also known as I_2. Melt index is inversely proportional to the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt index, although the relationship is not linear. The melt index for the substantially linear ethylene interpolymers useful herein is generally from 0.01 gram/10 minutes (g/10 min.) to 30 g/10 min., preferably from 0.1 g/10 min. to 20 g/10 min., and especially from 0.5 g/10 min. to 15 g/10 min.

Other measurements useful in characterizing the molecular weight of substantially linear ethylene polymers involve melt index determinations with higher weights, such as, for common example, ASTM D-1238, Condition 190 C/10 kg (formerly known as "Condition N" and also known as I_{10}). "Melt flow ratio" is defined herein as the ratio of a higher weight melt index determination to a lower weight determination, and for measured I_{10} and the I_2 melt index values, the melt flow ratio is conveniently designated as I_{10}/I_2.

Unlike heterogeneous conventional linear polymers which possess no substantial long chain branching, for the substantially linear ethylene polymers used to prepare the elastic films, strip, coatings, ribbons and sheets and elastic composite structures of the present invention, the melt flow ratio actually indicates the degree of long chain branching, that is, the higher the I_{10}/I_2 melt flow ratio, the more long chain branching in the polymer. The I_{10}/I_2 ratio of the substantially linear ethylene polymers is at least about 5.63, preferably at least about 6, and more preferably at least about 6.5. Generally, the upper limit of I_{10}/I_2 ratio for the homogeneous branched substantially linear ethylene polymers is about 50 or less, preferably about 30 or less, and especially about 20 or less.

The substantially linear ethylene polymers are analyzed by gel permeation chromatography (GPC) on a Waters 150 high temperature chromatographic unit equipped with differential refractometer and three

columns of mixed porosity. The columns are supplied by Polymer
Laboratories and are commonly packed with pore sizes of 10^3, 10^4, 10^5 and
10^6Å. The solvent is 1,2,4-trichlorobenzene, from which 0.3 percent by
weight solutions of the samples are prepared for injection. The flow rate
5   is 1.0 milliliters/minute and the operating temperature is 140 C with a
100-microliter injection size.

The molecular weight determination with respect to the polymer
backbone is deduced by using narrow molecular weight distribution
10   polystyrene standards (from Polymer Laboratories) in conjunction with
their elution volumes. The equivalent polyethylene molecular weights are
determined by using appropriate Mark-Houwink coefficients for polyethylene
and polystyrene (as described by Williams and Ward in <u>Journal of Polymer</u>
<u>Science</u>, Polymer Letters, Vol. 6, p. 621, 1968., to derive the following
15   equation:

$$M_{polyethylene} = a * (M_{polystyrene})^b.$$

In this equation, a = 0.4316 and b = 1.0. Weight average molecular
20   weight, M_w, is calculated in the usual manner according to the following
formula: $M_w = R w_i * M_i$, where w_i and M_i are the weight fraction and
molecular weight, respectively, of the ith fraction eluting from the GPC
column.

The M_w/M_n of the substantially linear homogeneously branched
25   ethylene polymers is defined by the equation:

$M_w/M_n \leq (I_{10}/I_2) - 4.63$. Preferably, the M_w/M_n is from 1.5 to 2.5,
- and especially about 2.

30   The homogeneously branched substantially linear ethylene polymers
can, however, have varying I_{10}/I_2 ratios due to long chain branch
incorporation, while maintaining a low molecular weight distribution
(i.e., M_w/M_n from 1.5 to 2.5).

The "rheological processing index" (PI) which is defined herein as the apparent viscosity in kpoise of a polymer measured by a gas extrusion rheometer (GER), can also be used to distinguish the substantially linear ethylene polymers of this invention. The gas extrusion rheometer is

5   described by M. Shida, R.N. Shroff and L.V. Cancio in Polymer Engineering Science, Vol. 17, No. 11, p. 770 (1977), and in "Rheometers for Molten Plastics" by John Dealy, published by Van Nostrand Reinhold Co. (1982) on pp. 97-99. GER experiments are performed at a temperature of 190°C, at nitrogen pressures between 250 to 5500 psig (17-379 bars) using about a

10   7.54 cm diameter, 20:1 L/D die with an entrance angle of 180°. For the substantially linear ethylene polymers described herein, the PI is the apparent viscosity (in kpoise) of a material measured by GER at an apparent shear stress of 2.15×10^6 dyne/cm^2 and is determined from the following equation:

15

PI = 2.15×10^6 dynes/cm^2/(1000 X shear rate),

where:

2.15×10^6 dynes/cm^2 is the shear stress at 2500 psi, and

20   the shear rate is the shear rate at the wall as
represented by the following equation:

32 Q'/ (60 sec/min)(0.745)(Diameter X 2.54 cm/in)^3,

25   where:

Q' is the extrusion rate (gms/min),

0.745 is the melt density of polyethylene (gm/cm^3), and

30   Diameter is the orifice diameter of the capillary (in.).

The unique substantially linear ethylene interpolymers and homopolymers described herein preferably have a PI in the range of 0.01 kpoise to 50 kpoise, preferably about 15 kpoise or less. The unique

35   substantially linear ethylene interpolymers and homopolymers described herein have a PI less than or equal to about 70 percent of the PI of a

comparative linear ethylene polymer (either a Ziegler polymerized polymer or a linear uniformly branched polymer as described by Elston in US Patent 3,645,992) at about the same I_2 and M_w/M_n.

5   To more fully characterize the rheological behavior of the unique substantially linear ethylene polymers, S. Lai and G.W. Knight recently introduced (ANTEC '93 Proceedings, INSITE™ Technology Polyolefins (ITP) - New Rules in the Structure/Rheology Relationship of Ethylene α-Olefin Copolymers, New Orleans, La., May 1993) another rheological measurement,

10   the Dow Rheology Index (DRI), which expresses a polymer's "normalized relaxation time as the result of long chain branching." DRI ranges from 0 for polymers which do not have any measurable long chain branching (for example, Tafmer™ products supplied by Mitsui and Exact™ products supplied by Exxon) to about 15 and is independent of melt index. In general, for

15   low to medium pressure ethylene polymers (particularly at lower densities) DRI provides improved correlations to melt elasticity and high shear flowability relative to correlations of the same attempted with melt flow ratios, and for the substantially linear ethylene polymers of this invention, DRI is preferably at least about 0.1, and especially at least

20   about 0.5, and most especially at least 0.8. DRI can be calculated from the equation:

$$\text{DRI} = (3652879 * \tau_0^{1.00649}/\eta_0 - 1)/10$$

25   where τ_0 is the characteristic relaxation time of the material and η_0 is the zero shear viscosity of the material. Both τ_0 and η_0 are the "best fit" values to the Cross equation,

$$\eta/\eta_0 = 1/(1 + (\dot{\gamma} * \tau_0)^{1-n})$$

30   where n is the power law index of the material, and η and γ are the measured viscosity and shear rate, respectively. Baseline determination of viscosity and shear rate data are obtained using a Rheometric Mechanical Spectrometer (RMS-800) under dynamic sweep mode from 0.1 to 100 radians/second at 160 C and a Gas Extrusion

35   Rheometer (GER) at extrusion pressures from 1,000 psi to 5,000 psi

(6.9 MPa to 34.4 MPa) (corresponding shear stress from 0.086 to 0.43
MPa) using a 7.54 centimeter diameter, 20:1 L/D die at 190 C.
Specific material determinations can be performed from 140 to 190 C
as required to accommodate melt index variations.

Additionally, ethylene polymers can also be distinguished by melt
flow consequences that are manifested as solid state surface defects such
as "melt fracture". An apparent shear stress versus apparent shear rate
plot is used to identify the "melt fracture" phenomena which pertains to
surface irregularities. According to Ramamurthy in the _Journal of_
Rheology, 30(2), 337-357, 1986, above a certain critical shear rate (in
contrast to a critical draw rate for the draw resonance phenomena), the
observed extrudate irregularities may be broadly classified into two main
types: surface melt fracture and gross melt fracture.

Surface melt fracture occurs under apparently steady flow
conditions and ranges in detail from loss of specular film gloss to the
more severe form of "sharkskin.." In this disclosure, the onset of surface
melt fracture (OSMF) is characterized at the beginning of losing extrudate
gloss at which the surface roughness of the extrudate can only be detected
by 40x magnification. The critical shear rate at the onset of surface
melt fracture for the substantially linear ethylene polymers is at least
50 percent greater than the critical shear rate at the onset of surface
melt fracture of a comparative linear ethylene polymer (either a Ziegler
polymerized polymer or a linear homogeneously branched polymer as
described by Elston in US Patent 3,645,992) having about the same I_2 and
M_w/M_n.

Gross melt fracture occurs at unsteady extrusion flow conditions,
and ranges in detail from regular (alternating rough and smooth, helical,
etc.) to random distortions. For commercial acceptability, surface
defects should be minimal, if not absent, for good film, coating or sheet
quality and properties. The critical shear stress at the onset of gross
melt fracture for the substantially linear ethylene polymers used in the
present invention is greater than about 4×10^6 dynes/cm^2. The critical
shear rate at the onset of surface melt fracture (OSMF) and the onset of

gross melt fracture (OGMF) will be used herein based on the changes of
surface roughness and configurations of the extrudates extruded by a GER.

The distribution of comonomer branches for substantially linear
ethylene polymers is characterized by its SCBDI (Short Chain Branch
Distribution Index) or CDBI (Composition Distribution Branch Index) and is
defined as the weight percent of the polymer molecules having a comonomer
content within 50 percent of the median total molar comonomer content.
The CDBI of a polymer is readily calculated from data obtained from
techniques known in the art, such as, for example, temperature rising
elution fractionation (abbreviated herein as "TREF") as described, for
example, by Wild et al., _Journal of Polymer Science, Poly. Phys. Ed._, Vol.
20, p. 441 (1982), or in US Patent 4,798,081. The SCBDI or CDBI for the
substantially linear interpolymers and homopolymers of the present
invention is preferably greater than about 30 percent, especially greater
than about 50 percent.

The substantially linear ethylene interpolymers used in this
invention essentially lack a measurable "high density" fraction as
measured by the TREF technique. The substantially linear polymers do not
contain a polymer fraction with a degree of branching less than or equal
to 2 methyls/1000 carbons. The "high density polymer fraction" can also
be described as a polymer fraction with a degree of branching less than
about 2 methyls/1000 carbons. One of the chief benefits of the lack of
high density polymer fraction is polymer materials with improved
stretchability and elasticity.

Additives such as antioxidants (for example., hindered phenolics
such as Irganox® 1010 or Irganox® 1076 supplied by Ciba Geigy),
phosphites (for example, Irgafos® 168 also supplied by Ciba Geigy), cling
additives (for example, PIB), Standostab PEPQ™ (supplied by Sandoz),
pigments, colorants, and fillers, although not required to achieve the
desired results of this invention, can also be included in the elastic
materials disclosed herein. However, the additives should be incorporated
in such manners or to the extent that they do not interfere with the
enhanced elasticity and stretchability discovered by Applicants. The

elastic films, strips, coatings, tapes, ribbons and sheets may also
contain additives to enhance antiblocking and coefficient of friction
characteristics. Still other additives, such as surfactants and
quaternary ammonium compounds alone or in combination with ethylene-
5   acrylic acid (EAA) copolymers or other functional polymers, may also be
added to effectuate desired wetting, antistatic or antimicrobial
characteristics of the elastic materials and elastic composite structures
of this invention.

10   Similar to the use of additives and although not required to gain
the benefits of the present invention, the substantially linear ethylene
polymers disclosed herein can be admixed with other polymers to modify
characteristics such as elasticity, processability, strength, thermal
bonding, or adhesion, to the extent that such modification does not
15   adversely affect the enhanced properties discovered by the Applicants.

         Some useful materials for modifying the substantially linear
ethylene interpolymers of this invention include, other substantially
linear ethylene polymers as well as other polyolefins, such as high
20   pressure low density ethylene homopolymer (LDPE), ethylene-vinyl acetate
copolymer (EVA), ethylene-carboxylic acid copolymers, ethylene acrylate
copolymers, polybutylene (PB), ethylene/α-olefin polymers which includes
high density polyethylene (HDPE), medium density polyethylene,
polypropylene, ethylene-propylene interpolymers, linear low density
25   polyethylene (LLDPE), ultra low density polyethylene (ULDPE), as well as
graft-modified polymers involving, for example, anhydrides and/or dienes,
or mixtures thereof.

         Still other polymers suitable for modifying the substantially linear
30   polymers of the present invention include synthetic and natural elastomers
and rubbers which are known to exhibit varying degrees of elasticity. AB
and ABA block or graft copolymers (where A is a thermoplastic endblock
such as, for example, a styrenic moiety and B is an elastomeric midblock
derived, for example, from conjugated dienes or lower alkenes),
35   chlorinated elastomers and rubbers, ethylene propylene diene monomer
(EDPM) rubbers, ethylene-propylene rubbers, and mixtures thereof are

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examples of known prior art elastic materials contemplated as suitable for modifying the elastic materials disclosed herein.

For the elastic composite materials of the present invention, any
5   layer (or portion) and any number of layers can comprise at least one substantially linear ethylene polymer. However, preferably, the elastic material disclosed herein comprises at least 10 weight percent of any mixture or composite.

10 The materials claimed herein are elastic. For the homogeneously branched substantially linear ethylene polymers used herein, the percent permanent set relates to polymer density or percent crystallinity for polymers selected at densities less than about 0.89 g/cm^3. Generally, the lower the density, the lower the permanent set as indicated in Figure 1.
15 For materials made from a homogeneously branched substantially linear ethylene polymer having a density less than about 0.89 g/cm^3, the percent permanent set is less than or equal to about 20 percent (that is, the recovery is at least about 80 percent).

20 Figure 1 also indicates at densities greater than 0.89 g/cm^3 up to 0.92 g/cm^3, the slope of the curve that describe the relationship between density and elasticity is surprisingly flat. Above 0.92 g/cm^3, as polymer density is further increased, the slope of the curve is steep indicating the type of predictable density/elasticity relationship generally known in
25 the art. The density/elasticity relationship of substantially linear ethylene polymers is also characterized by a steep slope in the preferred density range of less than about 0.89 g/cm^3. As such, Figure 1 is suitably labeled to indicate three relatively discrete regions of elasticity; the preferred range is assigned the "Elastic Region", the
30 intermediate range where the slope is unexpectedly flat is assigned the "Elasticity Transition Zone", and the final range is assigned the "Nonelastic Region".

Table 1 indicates with respect to comparative examples 7, 8, 13 and
35 14 that film prepared from a homogeneously branched substantially linear ethylene polymer having a density within the Elastic Transition Zone shows

less elasticity than heterogeneously branched linear ethylene polymers at equivalent or lower density and crystallinity. Moreover, Table 1 also indicates that the preferred homogeneously branched substantially linear ethylene polymers disclosed herein show superior elasticity and

5 stretchability in contradistinction to other homogeneously branched substantially ethylene polymers due to the lower density and crystallinity of the preferred materials. Surprisingly, as illustrated in Figure 2, preferred homogeneously branched substantially linear ethylene polymers are further distinguished from other homogeneously branched substantially

10 ethylene polymers due to their tendency to show enhanced elasticity as film thickness is reduced whereas other homogeneously branched substantially linear ethylene polymers where the polymer density is greater than about 0.89 g/cm^3 will show less elasticity at reduced film thicknesses.

15

Where the elasticity of the novel polymers of the present invention varies with respect to the thickness of the material, thicknesses less than about 22 mils, preferably from 0.1 mils to 20 mils, and more preferably, from 0.4 mils to 15 mils are considered to be within the

20 purview of this invention.

For the novel elastic materials disclosed herein, the melt index of the substantially linear polymer can be widely varied, with little impact on elasticity. This allows more design flexibility for elastic composites

25 and finished articles because the strength and retractive force of the elastic material can be changed independently of its elasticity. For example, the tensile strength properties of an elastic film can be changed by changing the polymer's melt index (decreasing the melt index increases the tensile strength properties), rather than by changing the thickness of

30 the film, thus permitting a better optimization of the "hand" (that is, feel) of an elastic composite fabric with the desired elastic/strength performance of the composite fabric. The melt index of the substantially linear ethylene polymers disclosed herein is limited in traditional ways as requirements respecting specific extrusion processes. As examples,

35 extrusion coating and injection molding processes typically require high melt indices to avoid excessive extrusion pressures and polymer shearing

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as well as to provide adequate melt flow characteristics, while blown film
processes generally require lower melt indices to achieve adequate bubble
stability.

5   The films, coatings, and sheets of the present invention may be
fabricated by any polyolefin extrusion technique known in the art,
including blown bubble processes, cast extrusion, injection molding
processes, thermoforming processes, extrusion coating processes, profile
extrusion, and sheet extrusion processes. Simple blown bubble film
10   processes are described, for example, in The Encyclopedia of Chemical
Technology, Kirk-Othmer, Third Edition, John Wiley & Sons, New York, 1981,
Vol. 16, pp. 416-417 and Vol. 18, pp. 191-192. The cast extrusion method
is described, for example, in Modern Plastics, Mid-October 1989
Encyclopedia Issue, Volume 66, Number 11, pages 256 to 257. Injection
15   molding, thermoforming, extrusion coating, profile extrusion, and sheet
extrusion processes are described, for example, in Plastics Materials and
Processes, Seymour S. Schwartz and Sidney H. Goodman, Van Nostrand
Reinhold Company, New York, 1982, pp. 527-563, pp. 632-647, and pp. 596-
602. The strips, tapes and ribbons of the present invention can be
20   prepared by the primary extrusion process itself or by known post-
extrusion slitting, cutting or stamping techniques. Profile extrusion is
an example of a primary extrusion process that is particularly suited to
the preparation of tapes, bands, and ribbons.

25   The elastic materials of the present invention can also be rendered
pervious or "breathable" by any method well known in the art including by
apperturing, slitting, microperforating, mixing with fibers or foams, or
the like and combinations thereof. Examples of such methods include, USP
3,156,242 by Crowe, Jr., USP 3,881,489 by Hartwell, USP 3,989,867 by
30   Sisson and USP 5,085,654 by Buell.

        Fabricated articles which can be made using the novel elastic
materials disclosed herein include composite fabric articles (for example,
disposable incontinence garments and diapers) that are comprised of one or
35   more elastic component or portion. For example, elastic components are
commonly present in diaper waist band portions to prevent the diaper from

falling and leg band portions to prevent leakage (as shown in USP
4,381,781 (Sciaraffa). Often, the elastic component promotes better form
fitting and/or fastening systems for a good combination of comfort and
security. The novel elastic materials disclosed herein can also produce
5   fabric composite structures which combine elasticity with breathability by
utilization of a technique that renders the elastic material pervious or
"breathable" such as suggested by Lippert et al. in USP 4,861,652 and
indicated above.

10   The novel elastic materials disclosed herein can also be used in
various structures as described in USP 2,957,512 (Wade). For example,
layer 50 of the structure described in USP '512 (that is, the elastic
component) can be replaced with the novel elastic materials, especially
where flat, pleated, creped, etc., nonelastic materials are made into
15   elastic or semi-elastic structures. Attachment of the novel elastic
materials to nonelastic or less-elastic materials can be done with heat
bonding or with adhesives. Gathered or shirred elastic composite
materials can be produced from the new elastic material described herein
and nonelastic components by pleating the non-elastic component (as
20   described in USP '512) prior to attachment, prestretching the elastic
component prior to attachment, or heat shrinking the elastic component
after attachment.

The recovery after heat shrinking can be further enhanced by
25   effectuating a high degree of orientation into the elastic material during
fabrication. Significant orientation can be accomplished by the
utilization of various known techniques such as high blow-up blown film
fabrication, tenter framing of cast films and "double bubble" or "trapped
bubble" blown film fabrication as described in copending U.S. application
30   serial number 08/055,063, filed April 28, 1993.

The novel elastic materials described herein can also be used make
other novel structures. For example, USP 4,801,482 (Goggans) discloses an
elastic sheet (12) which can now be made with the novel materials
35   described herein.

The novel elastic materials described herein can also be used to make breathable portion or breathable elastic composite materials. For example, USP 5,085,654 (Buell) discloses a leg band (15) with a breathable portion 45, a breathable topsheet (26), a breathable backsheet (25),

5 elastic elements (31 and 64), a breathable element (54), and a breathable sub-element (96) all or any combination of which can now be made with the elastic materials disclosed herein in either pervious or impervious forms.

The novel elastic materials disclosed herein also have adjustable

10 stretchability and elasticity that can be achieved by specific combinations of elastic materials and less-elastic material and/or by adjusting the polymer density or by specific combination of different substantially linear ethylene polymers, which enables design flexibility for variable stretchability or retractive force in the same garment, as

15 described for example in USP 5,196,000 (Clear et al.).

USP 5,037,416 (Allen et al.) describes the advantages of a form fitting top sheet by using elastic ribbons (member 12) and an elastic backsheet (member 16). Pervious novel elastic materials described herein

20 could serve the function of member 12 and impervious elastics materials of this invention could function as member 16, or disclosed elastic materials could be used in an elastic composite fabric form.

In USP 4,981,747 (Morman), the novel elastic materials disclosed

25 herein can be substituted for elastic sheets 12, 122 and 232 to construct an elastic composite material which includes a reversibly necked material.

Elastic panels, elements, portions or the like can also be made from the novel elastic materials disclosed herein, and can be used, for

30 example, as members 18, 20, 24, and/or 26 of USP 4,940,464 (Van Gompel). The novel elastic materials described herein can also be used, for example, as elastic composite side panels (for example, layer) or as elastic ribbons 42 and/or 44.

EXAMPLES

The following examples illustrate some of the particular embodiments of the present invention, but the following should not be construed to mean the invention is limited to the particular embodiments shown.

5

Inventive Examples 1-6 and Comparative Examples 7-14

Table 1 indicates the melting point, crystallinity and percent permanent set for the various films. Except comparative examples 13 and 14, all examples are comprised of a substantially linear ethylene/1-octene copolymer produced by a continuous solution polymerization in the presence of a constrained geometry catalyst system as described in pending U.S. application serial number 07/776,130, now U.S. Patent No. 5,272,236, and in pending U.S. application serial number 07/939,281, now U.S. Patent No. 5,278,272,. Comparative examples 13 and 14 are heterogeneous branched linear low density ethylene polymers.

10

15

Whereas examples 1-14 are cast films, Inventive Examples 15, 16 and 17 are blown bubble elastic films. Inventive Example 17 comprises 85 weight percent Inventive Example 1 and 15 weight percent of a low density polyethylene resin with a 0.7 g/10 min. Melt Index and a 0.9235 g/cm^3 density blended by tumble blending.

20

A three-extruder cast coextrusion film line equipped with a 20/60/20 feedblock design and a slot die was used to fabricate the films of Examples 1-14. The cast film line extruder diameters were 2-1/2, 3-1/2 and 2 inches (6.4, 8.9 and 5.1 cm), respectively, and the die width measured 76 cm with a 20-mil manual die gap. The fabrications consisted of maintaining melt temperatures at about 274 C to prepare 1.5 mil and 3.0 mil film samples.

25

30

Examples 15, 16 and 17 are blown film samples fabricated using a 2.5 inch (6.35 cm) diameter Gloucester extruder equipped with a 6 inch (15.2 cm) annular die. Output rate for the fabrication was maintained at 120 lb/hr. (54.5 kg/hr.) with the melt temperature at 440 F (227 C) and the blow-up ratio (BUR) at 2.5:1 to prepare from 0.75 mil to 2.5 mil layflat film.

35

The melting point and crystallinity of the resultant films were
determined using differential scanning calorimetry (DSC) analysis where
292 J/g was taken as the heat of fusion for a 100 percent crystallinity
5   polymer film.

The percent permanent set was obtained by cutting the films into 2.5
cm x 15 cm strips and testing each film on an Instron tensiometer equipped
with a small plastic jaw on the crosshead (the jaw had a weight of about
10   six grams) and a 500 gram load cell. The jaws were set 1 inch (2.54 cm)
apart. The cross head speed was set at 5 inches/minute (12.7 cm/minute).
A single film specimen was loaded into the Instron jaws for testing. The
film was allowed to return to the original Instron setting (where the jaws
were again 1 inch apart) and the film was again pulled. At the point
15   where the film began to provide stress resistance, the strain was recorded
and the percent permanent set was calculated. In one example, a film
pulled for the second time did not provide stress resistance (that is,
pull a load) until it had traveled 0.2 inches (0.5 cm). Thus, the percent
permanent set was calculated as 20 percent, that is, the percent of
20   elongation at which the film began to provide stress resistance. The
numerical difference between the percent permanent set and 100 percent is
known as the percent elastic recovery. Thus, a film having a permanent
set of 20 percent will have a 80 percent elastic recovery. After
recording percent permanent set, the film was pulled to 100 percent
25   elongation. The film pulling process was repeated five times for each
determination and repeated at least twice, average and recorded as percent
permanent set at the beginning of the fifth pull.

Examples 1-6 were all easily made into film and have less permanent
30   set (more elasticity) than comparative examples 7-14 due to their lower
density and crystallinity. Surprisingly, although comparative examples 7
and 8 showed a substantially lower density than comparative examples 13
and 14, the percent permanent set values for comparative examples 7 and 8
were either equivalent to or inferior to that of the heterogeneously
35   branched linear ethylene polymer. The elastic response of substantially
linear ethylene polymers in this range of densities ($0.89 g/cm^3$ to 0.92

g/cm^3) was surprisingly flat and could not be confidently predicted from polymer density. At all densities above about 0.89 g/cm^3, Table 1 indicates the materials were nonelastic (permanent set was greater than 20 percent, or recovery was less than 80 percent).

Table 1 also indicates that the elastic materials described herein tend to show enhanced elasticity as film thickness was reduced while substantially linear ethylene polymers above about 0.89 g/cm^3 density tend to show less or unchanged elasticity at reduced film thicknesses.

Table 1

Examples*	Melt Index g/10 min.	Density g/cc	MWD	I10/I2 Ratio	DSC Melting Point, C 1st Heat	2nd Heat	% Crystallinity 1st Heat	2nd Heat	Film Thickness (mils)	5th Pull % Permanent Set	% Standard Deviation
Inventive Example											
1	1.0	0.870	2.04	7.4	55.5	54.0	5.8	9.7	1.5	9.1	2.6
2									3.0	11.4	1.0
3	3.0	0.874	2.01	7.4	55.5	60.6	8.3	13.5	1.5	9.9	3.6
4									3.0	12.6	1.5
5	5.0	0.870	2.00	7.3	55.8	56.9	4.7	12.1	1.5	6.5	1.7
6									3.0	12.3	0.8
Comparative Example											
7	1.0	0.899	2.17	8.6	92.9	94.0	18.3	31.0	1.5	28.7	1.7
8									3.0	24.7	0.5
9	3.5	0.911	2.14	7.0	104.0	104.2	27.5	40.5	1.5	28.7	1.5
10									3.0	24.3	1.2
11	2.5	0.935	2.11	9.5	122.6	123.6	51.1	61.2	1.5	61.1	2.5
12									3.0	60.2	3.2
13	2.3	0.917	ND	7.4	123.6	121.8	31.6	45.2	1.5	27.3	1.5
14									3.0	29.5	0.9
Inventive Example											
15	1.0	0.870		7.4	ND	ND	ND	ND	1.5	6.6	2.2
16									4.0	12.3	1.5
17	–	–		–	ND	ND	ND	ND	5.0	12.8	0.5

*Permanent set and standard deviation are taken as average of machine and cross direction determinations. Melt points and %crystallinity are determined on films at 3 mils thickness.

ND = not determined.

Inventive Examples 15-17

Inventive Examples 15 and 16 indicate that there is no substantial
difference between fabricating the elastic materials disclosed herein by
cast extrusion or by blown film extrusion. Inventive Example 17 shows
5   admixtures comprised of up to 15 weight percent of other polyolefin (0.7
g/10 minutes melt index, 0.9235 g/cc density low density polyethylene)
does not adversely affect the excellent elasticity of the elastic
materials of the present invention.

10   Elastic Composite Material Example

In another investigation, an elastic composite structure 10 depicted
in Figures 3 and 3A was constructed according to the description in U.S.
Patent # 2,957,512. In this investigation, Inventive Example 6 herein was
substituted for layer 50 in Figure 9 of USP '512 and a lab-produced
15   nonwoven was substituted for layer 59 in Figure 9 of USP '512. Fibers for
the lab-produced nonwoven were prepared from ASPUN* 6811A fiber grade
resin (a heterogeneously branched ethylene/1-octene copolymer having a
density of 0.941 g/cm^3 and a melt index of 27 g/10 min. and was supplied
by The Dow Chemical Company) and spunbond by Polybond, Inc. to 1.5 oz/yd^2)
20   (0.05 kg/m^2). Whereas layer 50 in Figure 9 of USP '512 constitutes the
elastic layer, layer 12 herein in Figures 3 and 3A constitutes the elastic
layer, and whereas layer 59 in Figure 9 of USP '512 is the nonelastic
layer, layer 14 herein Figures 3 and 3A is the nonelastic layer. The
percent permanent set was 68 percent for nonelastic layer 14 herein.
25   After spunbonding, the fibrous nonwoven web was further processed by well
known thermal bonding calendering where the calender consists of a 5"
(12.7 cm) diameter smooth bottom roll and 5" (12.7 cm) diameter diamond
embossed top roll. Calendering conditions included a 400 psi (2.75 MPa)
nip pressure, a roll speed of ~4 rpm, and a ~200 F (93.33 C) roll
30   temperature. An elastic composite material 10 was formed by cutting or
sizing the calendered nonelastic nonwoven sheet into a 2 x 12 inch (5.08 x
30.48 cm) strip and cutting the 3 mil film of Inventive Example 6 into a 2
x 6 (5.08 x 15.24 cm) inch strip. The nonelastic nonwoven 14 was then
evenly pleated to create two evenly spaced folds and then at least one ply
35   of unstretched Inventive Example 6 elastic film was placed on top of
nonwoven. The construction was then sandwiched between Mylar™ polyester

film supplied by E.I. Du Pont de Nemours and Company to prevent sticking
to the heated rolls and inverted such that the elastic film was oriented
on the bottom of the construction. The construction was then passed
through the heated nip of the calender to melt bond the Inventive elastic
5   film to the nonelastic nonwoven and created three melt bond areas 20. The
resultant elastic composite fabric structure 10 exhibited good
stretchability where stretch-to-end performance was 139 percent and
percent elongation by Instron tensiometer determination was 600.

Claims:

1. An elastic material having a percent recovery of at least about 80 percent consisting essentially of at least one homogeneously branched substantially linear ethylene polymer characterized as having:

        a) a melt flow ratio, I_{10}/I_2, $\geq$ 5.63,

        b) a molecular weight distribution, M_w/M_n, defined by the equation: $M_w/M_n \leq (I_{10}/I_2) - 4.63$,

        c) a critical shear rate at onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear ethylene polymer having about the same I_2 and M_w/M_n, and

        d) a density less than about 0.89 g/cm^3,

wherein the elastic material is selected from the group consisting of films, strips, coatings, tapes, webs, ribbons, bands and sheets.

2. The elastic material of Claim 1 wherein the M_w/M_n of the homogeneously branched substantially linear ethylene polymer is less than about 3.5.

3. The elastic material of Claim 1 wherein the M_w/M_n of the homogeneously branched substantially linear ethylene polymer is from 1.5 to 2.5.

4. The elastic material of Claim 1 wherein the I_{10}/I_2 of the ethylene polymer is at least about 6.

5. The elastic material of Claim 4 wherein the I_{10}/I_2 of the ethylene polymer is at least about 7.

6. The elastic material of Claim 1 wherein the thickness of the material is less than about 22 mils.

7. The elastic material of Claim 1 wherein the ethylene polymer is an interpolymer of ethylene with at least one C_3-C_{20} α-olefin.

8. The elastic material of Claim 1 wherein the ethylene polymer is a copolymer of ethylene with a C_3-C_{20} α-olefin.

9. The elastic material of Claim 1 wherein the ethylene polymer is a copolymer of ethylene with 1-butene, 1-hexene, 4-methyl-1-pentene or 1-octene.

10. The elastic material of Claim 9 wherein the ethylene polymer is a copolymer of ethylene with 1-octene.

11. The elastic material of Claim 10 wherein the density of the ethylene polymer is from 0.85 to 0.89 g/cm^3.

12. An elastic composite material comprising at least one elastic material having a percent recovery of at least about 80 percent, wherein the elastic material consist essentially of at least one substantially linear ethylene polymer characterized as having:

a) a melt flow ratio, I_{10}/I_2, ≥ 5.63,

b) a molecular weight distribution, M_w/M_n, defined by the equation: M_w/M_n ≤ (I_{10}/I_2) - 4.63,

c) a critical shear rate at onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear ethylene polymer having about the same I_2 and M_w/M_n, and

d) a density less than about 0.89 g/cm^3,

wherein the elastic material is selected from the group consisting of films, strips, coatings, tapes, webs, ribbons, bands and sheets.

13. A fabricated composite article comprising at least one elastic material having a percent recovery of at least about 80 percent, wherein the elastic material consist essentially of at least one substantially linear ethylene polymer characterized as having:

a) a melt flow ratio, I_{10}/I_2, $\geq$ 5.63,

b) a molecular weight distribution, M_w/M_n, defined by the equation: $M_w/M_n \leq (I_{10}/I_2) - 4.63$,

c) a critical shear rate at onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear ethylene polymer having about the same I_2 and M_w/M_n, and

d) a density less than about 0.89 g/cm^3,

wherein the elastic material is selected from the group consisting of films, strips, coatings, tapes, webs, ribbons, bands and sheets.

14. A fabricated composite article comprising at least one elastic composite material, wherein the elastic composite comprises at least one elastic material having a percent recovery of at least about 80 percent, wherein the elastic material consist essentially of at least one homogeneously branched substantially linear ethylene polymer characterized as having:

a) a melt flow ratio, I_{10}/I_2, $\geq$ 5.63,

b) a molecular weight distribution, M_w/M_n, defined by the equation: $M_w/M_n \leq (I_{10}/I_2) - 4.63$,

c) a critical shear rate at onset of surface melt fracture of onset of surface melt fracture of a linear ethylene polymer having about the same I_2 and M_w/M_n, and

d) a density less than about 0.89 g/cm^3,

      wherein the elastic material is selected from the group consisting of films, strips, coatings, tapes, webs, ribbons, bands and sheets.
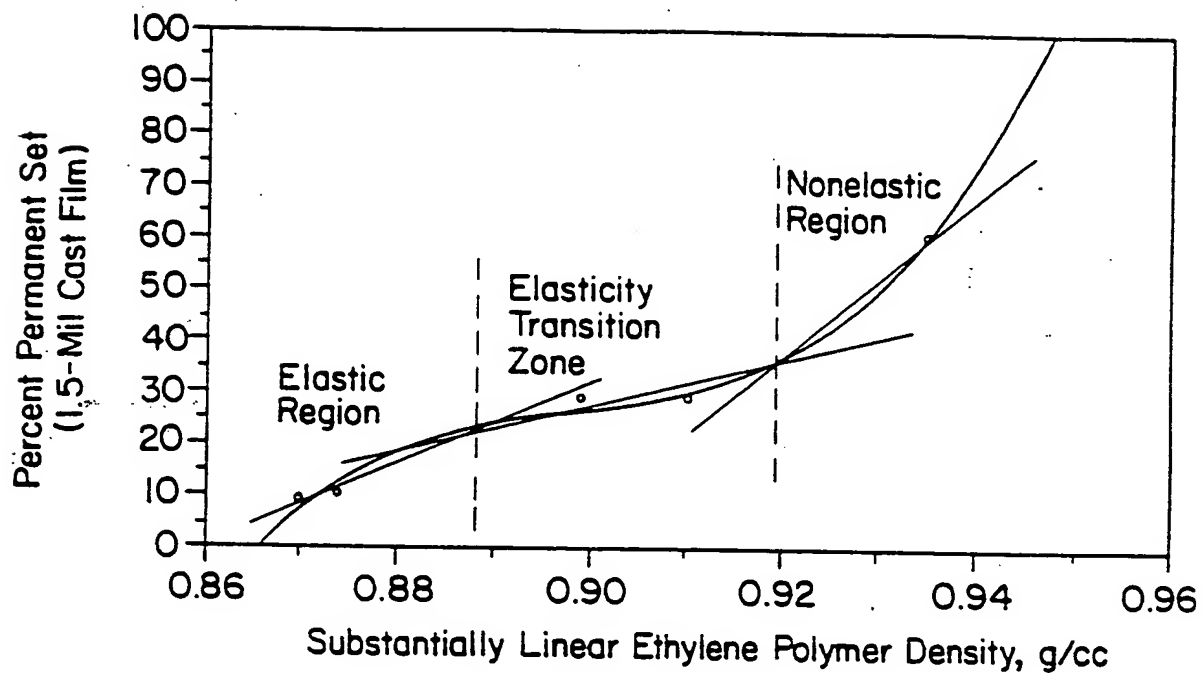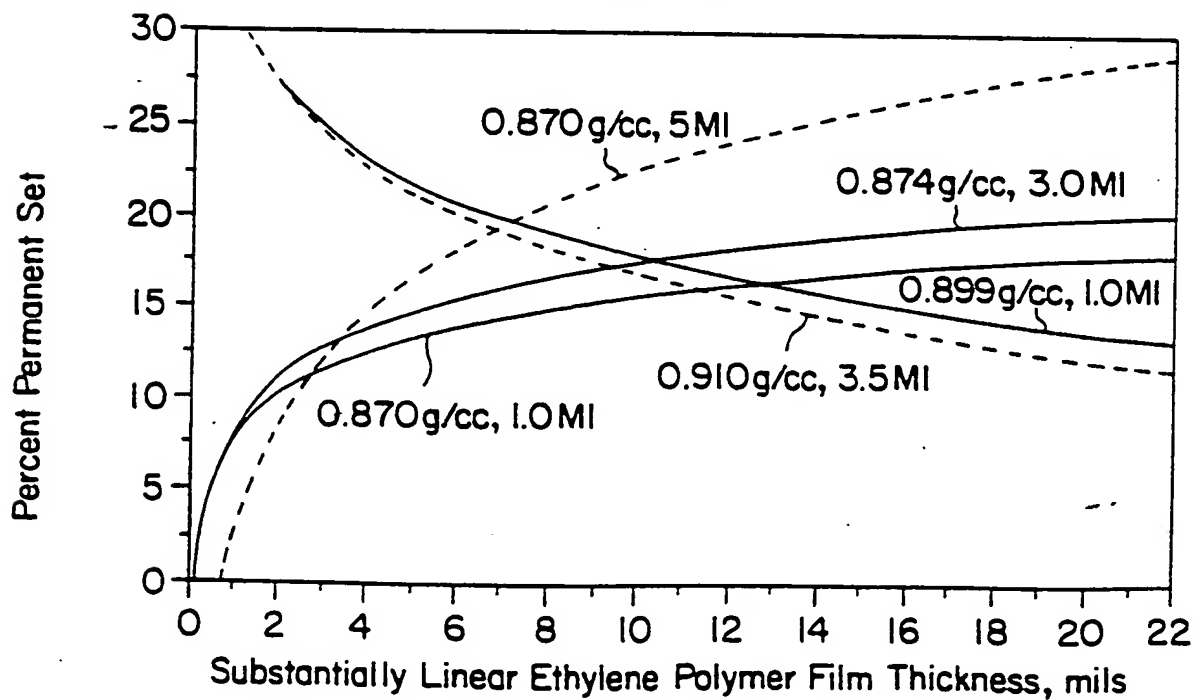
F I G. 1

F I G. 2

. CLASSIFICATION OF SUBJECT MATTER
IPC 6   C08L23/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6   C08L   C08J   C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO,A,93 08221 (THE DOW CHEMICAL COMPANY) 29 April 1993 see page 4, paragraph 2 - page 6, paragraph 2; claims see page 9, paragraph 3 - page 10, paragraph 1 see page 33, paragraph 3; example 24; table 10 -----	1-14

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

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Date of the actual completion of the international search	Date of mailing of the international search report
19 October 1994	11. 11. 94

Name and mailing address of the ISA	Authorized officer
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FIG. 3

FIG. 3A
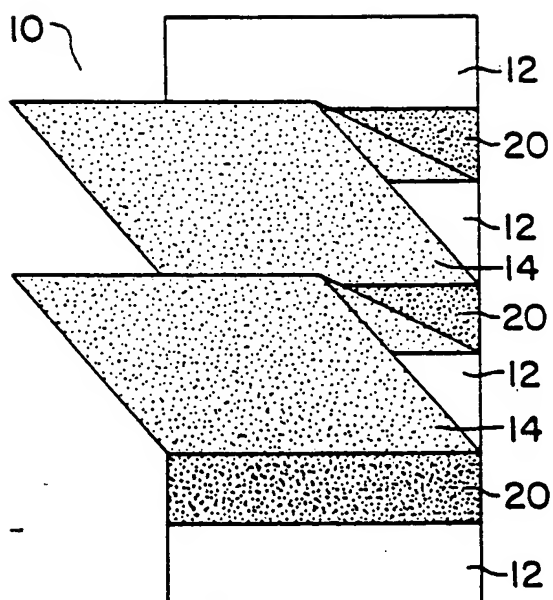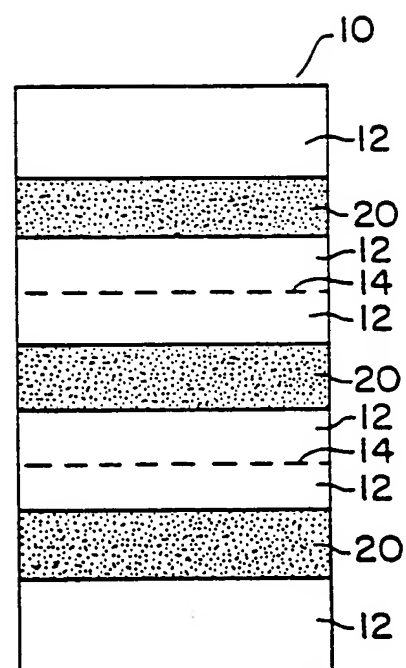
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